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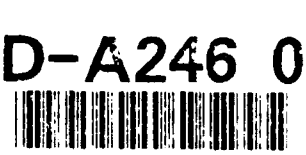

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pontaneous and Induced Coherent Radiation Generation In Atomic Vapors

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Professor Sven R. Hartmann

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The time delayed-four-wave-mixing technique using very wide band 00Å laser noise has been applied to study nile blue and relaxation imes in the low femtosecond regime have been measured as well as quantum eats which reveal vibronic structure. Relaxation in multi-level ystems has been analyzed and applied to the results in nile blue. ttosecond beats in K vapor have been observed and analyzed. Intererence effects have been observed in Na vapor which allowed the rasing of photon echo information. Linear and nonlinear effects of ocal field corrections have been analyzed and applied to third armonic generation and selective reflectivity. The local field orrections are found to effect both the line shape and the response requency. When the input fields are intense an analysis which goes

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beyond the slowly varying amplitude approximation reveals that the transverse profile of the input beam is modified.

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ULTRAFAST RELAXATION AND MODULATION IN NILE BLUE [1]

Time-delayed-four-wave-mixing, using incoherent laser pulses 5 ns long and 350 Å wide, has been used to measure the optical phase relaxation of a large molecule (nile blue) in solution at 300° K and 4.5° K. In the latter case an asymmetric response as well as a quantum beat (signaling a vibronic mode at 583 cm^{-1}) is observed. Its magnitude and shape gives its electron-vibron coupling factor $S = 0.79$ together with the homogeneous relaxation time T_2=43.7 fs and the inhomogeneous relaxation time T_2^*=13.3 fs (full width of 940 cm^{-1}) . Pump probe experiments [2] using 6 fs pulses show oscillations of such magnitude which can only be explained by invoking an S of comparable magnitude to that which we surmise.

ATTOSECOND BEATS AND MODULATION SPECTROSCOPY [3]

Two atomic states, dipole connected only to the ground state and not to each other, were each resonantly excited by a pair of temporally separated, noncolinear laser pulses. These pulse pairs were combined into two beams, each containing an undelayed component at one transition frequency and a delayed component at the other. The beams were arranged in the standard 2-beam degenerate four-wave mixing configuration. The four-wave mixing signal (measured at either frequency) then exhibited a periodic modulation as the time delay was varied. The frequency of this modulation (where frequency is understood as the inverse of the time-delay period) was either the sum or the difference of the two optical transition frequencies, depending on the input geometry.

Both potassium and sodium vapors were used as optical mixing media. In the case of potassium the excited resonances were the two fine-structure split 4S-2P levels. In the sodium the D-lines were excited. The sum-frequency modulation in potassium yielded the shortest time-delay period: 575 x 10^{-18} s or 575 attoseconds.

The study has also been extended to the regime in which the excitation field strengths exceed those for which perturbation theory, and hence the characterization of the mixing process as "four-wave" is valid.

This work is described in detail in Eric Usadi's PhD. thesis . A manuscript

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1. F. Moshary, M. Arend, R. Friedberg and S. R. Hartmann, Phys. Rev. Letters, submitted (1992)

2. H. L. Fragnito, J. Bigot, P. C. Becker and C. V. Shank, Chemical Physics Letters, **160**, 101 (1989)

3. E. Usadi, "Sum-Frequency and Difference-Frequency Modulation of Time-Delayed Four-Wave Mixing in Alkali Vapors", Thesis, Columbia University, New York, NY 10027, (1991)

is being prepared for publication in The Physical Review.

OPTICAL ECHO ERASING [4]

Photon echo interference effects have been used to selectively manipulate the amplitude and polarization of photon echoes generated by the read pulse in a long lived stimulated echo pulse sequence. The modulation depth of the interference is observed to decrease as the excitation beams are misaligned and no modulation is observed when the excitation beams are misaligned by more than a beam divergence. With regards to transient optical memory, this allows us to erase the information stored in randomly accessed bins of an optical memory device. This work is described in detail in Mark Arend's PhD. thesis. A manuscript is being prepared for publication in Optics Letters.

TIME DELAYED FOUR WAVE MIXING IN A MULTILEVEL SYSTEM [5]

The time delayed four wave mixing in a system of multilevel character such as nile blue is considerably more complicated than would be expected in an effective two level system such as found in an atomic vapor like Na [6]. To complement our experimental work in nile blue we studied the time-delayed-four-wave mixing response of an inhomogeneously broadened optical multilevel system with a single ground state. It was calculated for a pair of correlated excitation fields interrogated by a relatively coherent probe field. The effects of spectral diffusion (the sudden jump model [7]) were included and the longitudinal relaxation time of the excited multilevel system was taken to be short. With these specializations the response was found to be expressible in terms of elementary functions.

THIRD HARMONIC GENERATION IN GASES [8]

Three-photon resonantly enhanced third harmonic generation in a collisionally self-broadened gas of two-level atoms, appropriately buffered to allow phase matching, was found to achieve maximum output when the input frequency was sufficiently detuned from exact resonance so that excessive self-absorption of the third harmonic is avoided. For a given active gas density or laser detuning there exists a relation between the buffer gas density, active gas density and laser detuning which maximizes the third harmonic intensity. The optimal combination depends only on the physical characteristics of the

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4. M. Arend, "Photon Echo Interference Effects", Thesis, Columbia University, New York, NY 10027, (1991)
5. S. R. Hartmann and J. T. Manassah, Journal of Physics B (Atomic, Molecular and Optical Physics), **23**, 1363 (1990)
6. R. Beach, D. DeBeer and S. R. Hartmann, Physical Review A, **32**, 3467 (1985)
7. P. Hu and S. R. Hartmann, Physical Review B, **9**, 1 (1974)
8. R. Friedberg, S. R. Hartmann and J. T. Manassah, J. Phys. B: At. Mol. Opt. Phys., **24**, 2883 (1991)

tripling medium and the buffer gas. When the third harmonic is maximized the absorption coefficient α and the sample length L combine so that αL is of order unity. The characteristics of three-photon resonantly enhanced third harmonic generation in a buffered gas was analyzed in detail.

NONLINEAR EFFECT OF LOCAL-FIELD CORRECTION [9]

We developed a general formulation that incorporates local-field corrections into Bloch's equations and used it to calculate the strong-field susceptibility of a pressure-broadened gas consisting of two-level atoms. The major effect was found to be to introduce a dynamic Lorentz shift that varies throughout the resonance line shape. We also calculated the susceptibility associated with a weak field (probe) in the presence of a strong field (pump) when the pump and probe are at the same frequency. Here the effect was to introduce both a variable shift and a greater distortion of the line shape.

EFFECTS OF DYNAMIC LORENTZ SHIFT ON FOUR-WAVE PARAMETRIC INTERACTIONS [10]

We calculated the effect of the local-field correction (LFC) on the local response at ω_s and $2\omega_p - \omega_s$, of a medium consisting of two-level systems when subject to a pump of arbitrary intensity at ω_p and a weak probe at ω_s. The pump is considered strong if its reduced intensity $I = \omega_R{}^2 / \gamma_1 \gamma_2$ is large compared to unity. Here ω_R is the Rabi frequency and γ_1 and γ_2 are the longitudinal and transverse relaxation rates. In the weak pump case the LFC manifests itself as a fixed or stationary frequency shift. When the pump is strong, the response is independent of the LFC. At intermediate pump intensities the effect of LFC is dependent on the intensity of the pump. Both line-shift and line-shape distortions are present. The LFC thus gives rise to a shift in the resonance response that varies according to the intensity and frequency of the applied radiation.

DYNAMIC LORENTZ SHIFT AND REFLECTIVITY [11,12,13]

In a series of papers we proposed and analyzed the reflectivity response of a laser beam incident on a self-broadened gas. This response was found to be intensity dependent and able to display the dynamic Lorentz shift we had previously calculated [9].

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9. R. Friedberg, S. R. Hartmann and J. T. Manassah, Physical Review A, **40**, 2446 (1989)
10. R. Friedberg, S. R. Hartmann and J. T. Manassah, Physical Review A, **42**, 494 (1990)
11. R. Friedberg, S. R. Hartmann and J. T. Manassah, Opt. Comm., **75**, 363 (1990)
12. R. Friedberg, S. R. Hartmann and J. T. Manassah, Physical Review A, **42**, 5573 (1990)
13. R. Friedberg, S. R. Hartmann and J. T. Manassah, Journal of Physics B (Atomic, Molecular and Optical Physics), **24**, 3981 (1991)

SPATIAL MODIFICATION OF A GAUSSIAN BEAM ON REFLECTION FROM A SATURABLE ABSORBER [14]

We calculated the reflection coefficient of a dielectric-gas interface including the nonlinear local field, saturation, and self-reflected wave effects. We specialized to collisionally broadened gases with absorption lengths that are necessarily small compared with an optical wavelength and therefore preclude the use of the slowly varying amplitude approximation. The dependence of the reflectivity on the intensity of the incoming field is revealed through the changes that it induces in the transverse profile of the reflected Gaussian beam. These changes derive principally from effective aperturing effects in amplitude and phase.

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14. S. R. Hartmann and J. T. Manassah, Optics Letters, **16**, 1349 (1991)